

A setup for grazing incidence fast atom diffraction

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Peng Pan, Jaafar Najafi Rad, and Philippe Roncin^{a)}

AFFILIATIONS

Université Paris-Saclay, CNRS, Institut des Sciences Moléculaires d'Orsay (ISMO), 91405 Orsay, France

^{a)}Author to whom correspondence should be addressed: philippe.roncin@universite-paris-saclay.fr

ABSTRACT

We describe a UHV setup for grazing incidence fast atom diffraction (GIFAD) experiments. The overall geometry is simply a source of keV atoms facing an imaging detector. Therefore, it is very similar to the geometry of reflection high energy electron diffraction experiments used to monitor growth at surfaces. Several custom instrumental developments are described making GIFAD operation efficient and straightforward. The difficulties associated with accurately measuring the small scattering angle and the related calibration are carefully analyzed.

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I. INTRODUCTION

Discovered independently in Orsay¹ and then in Berlin,² grazing incidence fast atom diffraction (GIFAD) at crystalline surfaces has developed as a powerful analytic tool in surface science (see Refs. 3 and 4 for reviews). It is a grazing incidence version of thermal energy atom scattering (TEAS), just as reflection high energy electron diffraction (RHEED) is the high energy and grazing angle version of normal incidence low energy electron diffraction, LEED. GIFAD provides detailed information on the crystallographic structure of the topmost layer, atomic positions,⁵ and electron density.^{6,7} It does not induce defects or charging of the surface. The decoherence due to thermal motion is reduced mainly because the average position of the atomic rows has a larger effective mass⁸ than the constituting atoms, allowing for operation at elevated temperatures, as illustrated in the schematic view in Fig. 1. Together with the grazing geometry leaving the volume above the surface free for evaporation cells, GIFAD is well suited for molecular beam epitaxy (MBE).⁹ It can also count the number of deposited layers.¹⁰ GIFAD is a flexible technique that demonstrates diffraction on metal surfaces,^{11,12} semiconductors,^{6,13} and insulators.^{14–16} It also provided clear diffraction from mono-atomic oxide^{15–17} or molecular layers grown *in situ*,^{18,19} and immediately reveals the moiré modulation of a deposited graphene layer.^{20,21} It is completely insensitive to electric or magnetic fields.

The general GIFAD setup is presented in Fig. 2. It consists of an ion source combined with a neutralization cell, two divergence-limiting slits or diaphragms, a vacuum chamber holding the sample

surface, and an imaging detector.²² All these elements will be discussed in Secs. II–IV both from principle and in view of practical implementation, trying to evaluate what new idea worked efficiently or turned out to be problematic or useless. Section V is entirely devoted to the detector system and the corrections of its aberrations. The different modes of operation, θ -scan, ϕ -scan, E -scan, T -scan, and time-scan, are described in Secs. VII–IX to illustrate typical applications. In Sec. X, we discuss additional equipment taking benefits from the ion or atom beam together with the imaging detector inside the UHV chamber. We also discuss the use of miniature pointing lasers to help face problems specific with small angles geometries. Concluding remarks are given in Sec. XI.

II. THE ION SOURCE

As will be detailed below, GIFAD requires an atom beam injected through tiny diaphragms to reduce its divergence. In other words, the brightness is most important, and it requires a limited energy dispersion of the ion source. This can later be limited not only by the ionization mechanism but also by the geometry and intensity of the extraction field. The maximum current is expressed in μA corresponding to $\sim 5 \times 10^{12}$ ions per second, while only a few 10^3 – 10^4 atoms per second, but having the best resolution, will be needed to record a diffraction image in a few seconds.

A. Filament ion source

Hot filament ion sources have the reputation of providing bright ion beams in the keV energy range as required for

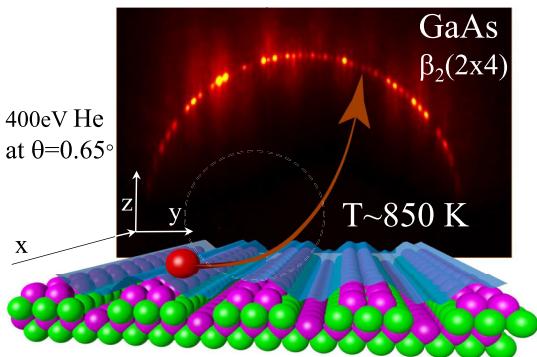


FIG. 1. Schematic view of GIFAD. The atomic wave of a projectile impinging at grazing incidence θ diffracts along the atomic row of the GaAs target surface (taken from Ref. 6). The image of the scattered particle shows well-defined diffraction spots sitting on a circle associated with energy conservation. The blue sheet symbolizes the iso-energy surface where the projectile is reflected.

spatially resolved secondary ion mass spectroscopy (SIMS) or depth profiling²³ applications. We used the EX05 model from VG Ortec with a differential pumping port, allowing for a beamline vacuum of better than 5×10^{-7} mbar. The EX05 is equipped with two electrostatic lenses and deflectors, providing flexible adjustment of the beam size, position, and focal distance. Note that a specific model EX05-N was developed for SIMS or (Auger electron spectroscopy) AES with a neutral beam option, but it is not commercially available anymore. We also tested a non-sequitur-technologies® ion gun,²⁴ also designed for SIMS applications, which significantly increases beam intensity but does not offer co-axial optical access. Most ion guns designed for focused ion beams and, therefore, equipped with ion optics should provide alternate solutions.

B. ECR ion source

Electron cyclotron resonance (ECR) ion sources use a magnetic structure and microwave to heat a plasma. They do not have a filament and can offer very long continuous operation and the ability to ionize any material either introduced directly as a gas or as a bulk material indirectly heated inside the source. GIFAD was initially discovered on a setup designed with Morosov from IPM Moscow²⁵ to investigate grazing incidence scattering of ions on surfaces. Then, it was equipped with a 10 GHz, NanoGan²⁶ Electron Cyclotron Resonance (ECR) ion source designed to deliver μA of Ar⁸⁺ ions. It

operates at low pressure ($P \sim 10^{-7}$ mbar), and the ionization efficiency is so significant that it acts as an ion pump with the vacuum inside the source decreasing significantly when the extraction field is turned on. Many different charge states were present in the source, so a high-performance magnet was needed to select the ion charge state of interest, resulting in a comparatively long beamline. We also used a high pressure ($P \sim 10^{-3}$ mbar), TES-40 ECR ion source from Polygon Physics²⁷ to produce atomic or molecular ion beams of hydrogen.

C. The Wien filter

A mass filter is mandatory when operating the ECR ion source with molecular gas such as H₂ to produce H⁺ or H₂⁺ ions, but we did not use it when working with high purity noble gas from the hot filament ion sources. We used a commercial Wien filter from non-sequitur-technologies® where the permanent magnet is placed outside the vacuum and can be removed, for instance, for baking purposes. For a more compact setup, a custom Wien filter taking advantage of the narrow collimation needed for GIFAD can probably be designed.

III. THE CHARGE EXCHANGE CELL AND BEAM COLLIMATION

The charge exchange cell is a $l = 2$ cm long tube with entrance and exit holes designed so that the internal pressure can be adjusted in the 10^{-3} mbar range without a dramatic increase of the background pressure in the beamline. For the resonant neutralization of 1 keV He⁺ ions on helium, a total cross section of $\sigma \approx 10^{-15} \text{ cm}^2$ was measured together with a mean scattering angle of around 0.1° at 1 keV.²⁸ The optimum pressure is calculated such that single collisions dominate, i.e., such that the probability $P = \sigma n l$ for an He⁺ ion to capture an electron from the target gas of the cell is around 10%, yielding a target density of $n \approx 5 \times 10^{13}$ particle per cubic cm, i.e., a pressure inside the cell around 5×10^{-4} mbar. The cell is closed at both ends by fixed holes ≈ 1 mm in diameter. In front of these holes, a linear translation sealed with a hydro-formed bellows brings a series of five fixed pinholes in clear view of the coarse 1 mm hole. The entrance hole is often left at maximum diameter, while the exit pinhole called \mathcal{O}_1 is used as the first divergence limiting diaphragm. The second one \mathcal{O}_2 is placed at a distance $L = 0.5$ m downstream, at the end of an injection tube, prolonging the beamline inside the UHV chamber until a few cm before the target. The tube also ends with a coarse $\mathcal{O} = 1$ mm hole, ensuring differential pumping. Another system holding a series of 12 pinholes made by electro-erosion is positioned on a miniature DN10CF rotation feed-through. For this system, where more room is available, the pinhole sizes defining \mathcal{O}_2 range from 10 to 200 μm by relative steps of $\sqrt{2}$, allowing for an area change by successive factor two; six additional vertical and horizontal slits are also present to allow sheet-shaped beams either parallel or perpendicular to the surface. For the circular holes, the resulting angular definition is $\delta\theta = (\mathcal{O}_1 + \mathcal{O}_2)/2L$ that can be adjusted down to 2×10^{-4} rad ≈ 1 mdeg. The transverse energy spread of a beam of energy $E_0 = 1$ keV is $E\delta\theta^2$, which can be as low as 0.5 meV, measured as the width of bound states resonances on the LiF surface.²⁹ Needless to say that the intensity passing through both diaphragms is severely reduced. For most experiments, we use diaphragms between

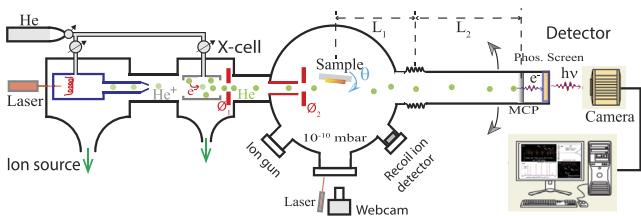


FIG. 2. Sketch of the GIFAD setup with four main parts: ion source, neutralization cell, UHV chamber, and detector system.

20 and 50 μm . The pinholes are separated by a distance of 1.2 mm, corresponding to an angle of $\delta\alpha = 2.7^\circ$, which is very difficult to read accurately on a miniature rotary drive. We had to implement a more accurate electronic readout using a tiny magnet coaxial to the rotary drive so that its absolute angular position can be read without contact by an AS5140 magnetic sensor. It is read by using an Arduino-mini micro-controller updating a four-digit LED display. Rather than the absolute angular position α in degree, we chose to display it as a four-digit float $\text{Pos} = (\alpha_i - \alpha_0)/\delta\alpha$ where the integer part identifies the pinhole number and the remainder is the actual position of the pinhole in front of the $\varnothing = 1$ mm diameter. For instance, a value of $\text{Pos} = 10.12$ display indicates diaphragm number 10, but positioned 0.12 pinhole separation units above the center of the coarse hole. Note that the fractional value is important since the exact position of the pinhole affects the actual beam position. Figure 3 shows the charge exchange cell designed to be mounted on a 63CF flange by an insulator so that it can be floated to change the energy of the ion beam before charge exchange without de-tuning the Wien filter placed in front. This allows for an ion beam transport at keV energy and deceleration immediately before neutralization. On both sides of the gas cell, two complete sets of deflectors are positioned co-axially on stacked 2 mm thick plates separated by thin Kapton disks. Each plate has a 0.8 mm hole drilled on its edge, which serves as a female connector for UHV twist pin connectors having their own spring. The deflectors before the cell were intended to steer both the position (y, z) and the direction (θ_y, θ_z) of the ion beam before entering the cell, but, in practice, mainly the angular control is used. The ones placed after the cell do not affect the neutral beam and are used to deflect the residual ion beam away. Additionally, when connected to a voltage pulse generator, these deflectors can produce a pulsed ion beam useful for surface contaminant analysis, as detailed in Sec. X B. In addition, it is important to be able to mis-align the ion source so that neutral atoms produced at the extraction level and having ill-defined energy do not contribute. This can be achieved by forcing mechanically the beamline to be tilted by $\sim 1^\circ$ or by positioning the diaphragms slightly off-axis until this signal disappears and then steering the ion beam inside the cell and experimenting with deflectors.

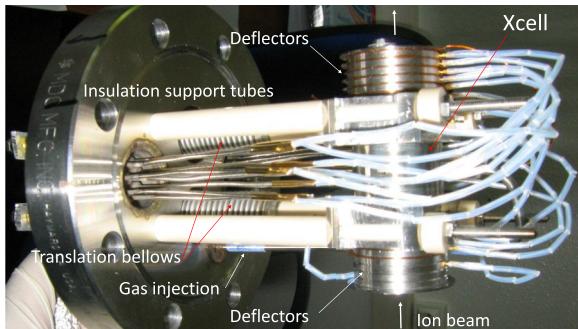


FIG. 3. The neutralization cell in the center is surrounded by two sets of two horizontal and vertical deflectors spot-welded on stacked disks separated by Kapton foils. The two bellows are to select and position the entrance and exit diaphragms. Reproduced with permission from P. Soulisse, “Développement d'un dispositif expérimental pour la diffraction d'atomes rapides et étude de surfaces d'isolants ioniques,” Ph.D. thesis, Université Paris Sud 11, 2011.³⁰

As described above, the pressure inside the charge exchange cell lies in the 10^{-3} mbar range, and the entrance and exit diaphragms limit the gas flow so that the pressure just around the cell is below a few 10^{-7} mbar. The coarse 1 mm hole connecting to the UHV chamber limits the variation of the pressure inside the UHV chamber below 10^{-9} mbar. If an additional 60 l/s pump and a coarse 1 mm hole are placed in between, the pressure of a few 10^{-10} mbar inside the UHV chamber is not at all affected by the gas introduced in the ion source or in the gas cell. The high pressure limit for the use of GIFAD is probably around 10^{-4} mbar where the mean free path for elastic scattering becomes less than a few cm. All the turbo-pumps of the beamline and introduction chamber are connected to a single dry pump.

IV. THE UHV MANIPULATOR

GIFAD is a diffraction technique and, therefore, requires accurate control of two angles, the angle of incidence of the atomic beam θ and the orientation of the surface ϕ . More precisely, $\theta = \pi/2 - \angle(\vec{k}_{in}, \vec{S})$ and $\phi = \pi/2 - \angle(\vec{k}_{in} \times \vec{S}, \vec{u})$, where the vectors \vec{k}_{in} , \vec{S} , and $\vec{u}_{h,k,0}$ indicate, respectively, the beam direction, the surface normal, and the surface crystallographic axis labeled by its Miller indices h, k .

A simple manipulator is characterized by a holding flange with a three-axis X, Y, Z translation stage holding a primary rotation axis perpendicular to this flange and terminated by a sample holder. More sophisticated models support a co-axial translation or rotation mechanism to perform additional movements, in our case an additional rotation. We have used two distinct configurations sketched in Fig. 4, where the high precision primary rotation axis is affected to the angle of incidence θ or to the azimuthal angle ϕ . In Fig. 4(a), the additional rotation involves an in-vacuum gear system, allowing for unlimited ϕ movement, whereas in Fig. 4(b), a separate ± 10 mm movement of the support flange translates into a variation of $\pm 4^\circ$ of the angle of incidence. The accuracy is excellent, but a modification of θ is associated with a significant shift of the target position mainly along the x direction affecting the angular calibration.

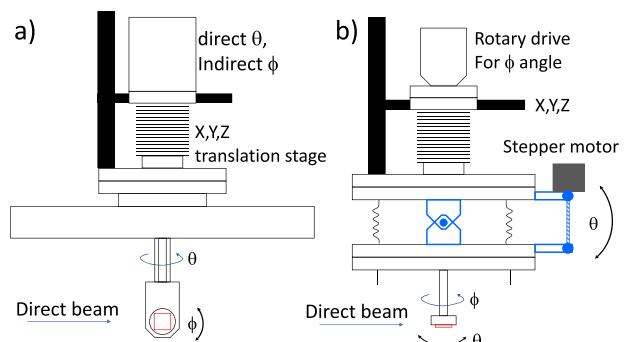


FIG. 4. Two different manipulators have been used, both fitted on a X, Y, Z platform. (a) Two coaxial rotations: one direct for θ and one indirect for ϕ . (b) Only one rotation assigned to ϕ ; the parts in blue have been welded to a DN100CF bellow so that θ is controlled by changing the parallelism of the flanges.

This is not the case with the manipulator MC from the UHV-design in Fig. 4(a); in the vacuum, azimuthal movement is not precise enough for crystallographic applications.

When operating inside a MBE chamber at INSP,^{6,9,10} the local manipulator was equipped with complete azimuthal freedom but limited control of the angle of incidence relative to the beam, so we decided to tune the angle θ by changing the beam direction. The ion source and neutralization cell were placed on a motorized bench connected to the MBE chamber by a UHV valve and flexible bellows. The combined control of two motors, one placed close to the ion source and the other one after the neutralization cell, was used to create a virtual center of rotation at the center of the target wafer in the MBE chamber.

One of the main interests of GIFAD is its compatibility with the MBE environment where it can track online the transformation of the topmost layer with temperature and the growth mode of the grown layers and their specific crystalline structures. In this respect, a sample holder having the ability to bring the sample at elevated temperatures or to low temperatures was mandatory. In studying thermal effects in inelastic diffraction on a LiF surface, we could explore temperatures between 140 and 1017 K³¹ where the limitation is given by the noise on the detector, probably due to electronic and ionic emission from the filament.

V. THE DETECTOR SYSTEM

A. Detecting neutral atoms

Detecting a neutral atom at thermal energy is not straightforward; it carries, on average, only $E = kT = 25$ meV kinetic energy and is not able to extract an electron when hitting a surface and cannot, therefore, be detected by using micro-channel-plates (MCPs). For helium, the only solution is to ionize it, and since the ionization potential is 24.58 eV, this is achieved efficiently only by electron impact. The process is not selective and also ionizes the residual gas so that helium atom detection is most often associated with an additional mass selective procedure, for instance, a quadrupole mass spectrometer. This is a major limitation of TEAS where angular resolution requires the use of small apertures, whereas analyzing several diffraction orders requires scanning the detector or sample over a large angular range.

The situation is much more favorable at keV energies where the atoms can be detected with a quantum efficiency of above 10%.³² When a keV atom tries to penetrate a solid, atomic collisions with surface atoms take place at close internuclear distances so that the overlap of the compact helium ground state electrons will push away the valence electrons of the target atom. The electrons ejected from the surface can be accelerated and “multiplied” until detection. Here, multiplication relies on the fact that an electron with a few hundred eV energy impacting specific materials will result in the emission of, on average, more than two secondary slow electrons. These are the processes at work in a photomultiplier tube where after impacting ten plates, a burst of $\approx 10^7$ count electrons is produced in less than a ns, producing a voltage peak of a few tens of mV on a 50 Ω resistor. In a Channeltron, all these ≈ 10 successive electronic impacts occur along the tube because it is curved, while in micro-channel-plates (MCPs), they occur inside each pore because of its very long aspect ratio $L/\phi_{pore} \approx 50$, where L and ϕ_{pore} are the pore length and

diameter, respectively. The gain of a single tube is limited to a few thousand electrons, but the use of two stacked MCP allows for a gain of $\approx 10^6$ electrons per impact.

B. Imaging detectors

Imaging detectors are particle detectors with the ability to localize the (x, y) coordinates of the impact of the particle within the detector input area. This is comparatively simple with MCP, where each pore can be seen as a Channeltron tube, providing millions of independent detectors. Knowing exactly which pore has been hit is possible but still difficult. Several compromises have been proposed (see, e.g., Ref. 33). One approach is to use dedicated electrodes and electronics, and this requires the use of two stacked MCP because the gain of a single MCP is limited by space charge to a few 10^3 electrons. The two most popular families are either charge division techniques, such as continuous resistive anodes,¹ which provide good differential linearity but poor integral linearity, or delay line anodes, allowing for a larger count rate and high resolution² and integral linearity, but with a limited differential nonlinearity requiring specific calibration.³⁴ Even without impact, the detector may trigger randomly, but this can be limited to only a few dark events per second randomly distributed on the MCP active surface so that the signal-to-noise ratio is usually very large. The other option we have adopted is to accelerate the electrons of the MCP cloud onto a phosphor screen and image the fluorescence with a camera.³⁵ This combination of MCP and phosphor screen, developed initially for military night vision systems, is technologically mature and offers excellent performances in terms of count rate, spatial resolution, and uniformity as detailed below. We use a single 80 mm MCP placed 1 mm ahead of a standard 3 mm thick glass plate coated with an indium tin oxide (ITO) layer and a 4 μm P43 phosphor over-layer. The light conversion efficiency is around 20 eV of electron energy per emitted photon so that a 2 kV bias between MCP and screen allows each of the 10^3 – 10^4 electrons of the MCP to produce ≈ 100 green photons, enough to detect a single impact with a proper lens and camera (see Sec. V D). The screen is stable under UHV conditions and can resist temperatures of up to 200 °C without losing performance.

We did not carefully investigate the problem of MCP uniformity, hoping that our $\sigma = 50$ μm spot size covering at least ten adjacent pores ($\approx (\sigma/\phi_{pore})^2$) would average single pore defects. We did not experience dark spots or gain variation except at the molecular beam epitaxy chamber, where exposure to gallium and arsenic together with the helium impacts were suspected to be responsible for reducing the efficiency in the central zone of the detector.

C. Position, shape, and intensity aberrations of the lens

Accurately measuring the scattered positions and intensities is the basis of any diffraction techniques, but this is crucial in GIFAD because of the small values of the scattering angles. As an example, the peak separation in Fig. 1 is around 0.05°. We analyze here the distortions introduced by the lens system imaging the phosphor screen through a UHV window at a distance of 25 cm. The lens is a high aperture F0.95 “xenon” lens from Schneider³⁶ with a focal length of 17 mm; it is operated with its diaphragm wide open. We

evaluated that a spot at the edge of the detector hits the lens with a maximum angle below 10° . This choice of the lens and camera position derives from a compromise between light collection efficiency and optical aberrations. We use the pixel of our (1920×1440) digital camera as a unit. The aberration of the lens has been tested *in situ* by imaging a fixed beam at different detector position and *ex situ* by scanning a light spot or imaging a rectangular grid. We determined the correspondence between the actual and measured beam position and track the evolution of the Gaussian width σ displayed in Fig. 5. The defects can be neglected in the central (paraxial) region but can reach almost half a millimeter error in the spot position at the edge. This error is easily corrected by a single parameter describing a cubic barrel-type aberration of Eq. (1), where ρ and ρ_c are the measured and corrected radial coordinates or distance to the center of the sensor expressed in pixel and a is the measured coefficient. Within present settings, the maximum distance to the center can be close to 800 pixels, yielding an error of ten channels. After this cubic correction, each pixel corresponds to its ideal location on screen with an error less than a pixel ($\approx 50 \mu\text{m}$ with our magnification) over the whole screen. The spot width was found approximately constant, while the skewness evolved linearly from -0.25 to 0.25 across the whole diameter.

Equation (2) reports the correction of the measured intensity deriving from the Jacobian of the position correction in Eq. (1). Finally, Eq. (3) reports the quadratic correction compensating for the vignetting aberration describing the radial intensity decrease; σ_v is the measured range in pixel units, indicating an $\sim 25\%$ drop at the position corresponding to the edge of our detector. With these simple polynomial radial forms, the position and intensity corrections are performed on the fly in our analysis software,

$$\rho_c = \rho(1 + a\rho^2), \quad a = 2.0 \times 10^{-8}, \quad (1)$$

$$I_c = I/(1 + 4a\rho^2), \quad (2)$$

$$I_v = I_c(1 + \rho^2/2\sigma_v^2), \quad \sigma_v = 1080. \quad (3)$$

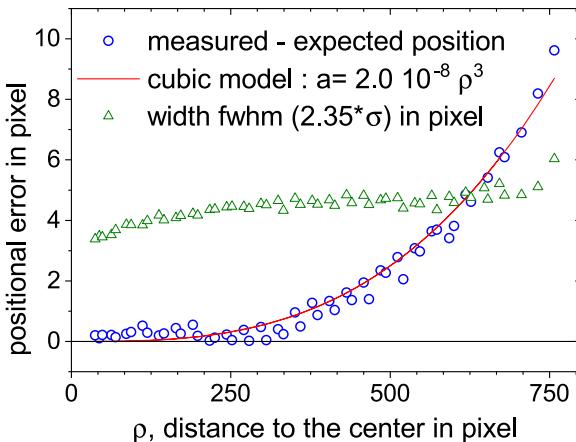


FIG. 5. The lens aberration, measured as the spot position on the camera minus its actual value measured with a caliper, is well fitted by a cubic coefficient a [Eq. (1)]. The spot width is approximately constant. The intensity was found to decrease with a quadratic character.

D. The camera aberrations

The other aberration observed is related to the camera itself, in our case, a Hamamatsu C11440 ORCA Flash, CMOS. We have noted that the image of a sharp spot is, indeed, a quasi-delta limited by the lens resolution, but this delta is sitting on top of a broad circular pedestal decaying slowly from the center to an edge with an almost 50 pixel radius. Its magnitude immediately at the foot position is only a fraction of a percent, but its integrated intensity is close to 25%. This was not anticipated since we switched from CCD to CMOS technology to avoid the blooming and charge leaking problems. The observed pedestal does not depend on the exposure time, so we believe that it is due to light scattering in the microlenses covering each pixel to achieve a very large light collection efficiency.

The problem could be corrected by software, but we decided to operate the camera in the single-particle detection mode, which requires that each impact produces enough photons to form a detectable spot in the image. To optimize this mode, we raised the MCP bias voltage to 1100 V so that each atom impact generates an estimated number of electrons close to 10^4 , each one producing almost 100 photons on the phosphor screen (P43 spec is 20 eV electron energy per emitted photon). Only 0.04% of these 10^6 photons will be collected by our lens for each impact, but this is significantly above the background level. The HiPic camera software provided by Hamamatsu offers a so-called “Photon counting mode” perfectly suited for this mode of operation.

In each image, the spots are searched, and a centroid detection is performed. Each location where the spot has been detected with an intensity above a given threshold is filled with a one, while other pixels are set to zero so that the camera behaves like a single particle detector, providing a valuable instant count rate. The detector also becomes insensitive to moderate stray light because a single photon on the screen is not enough to pass the threshold. The software embedded inside the camera can sustain such a spot analysis with

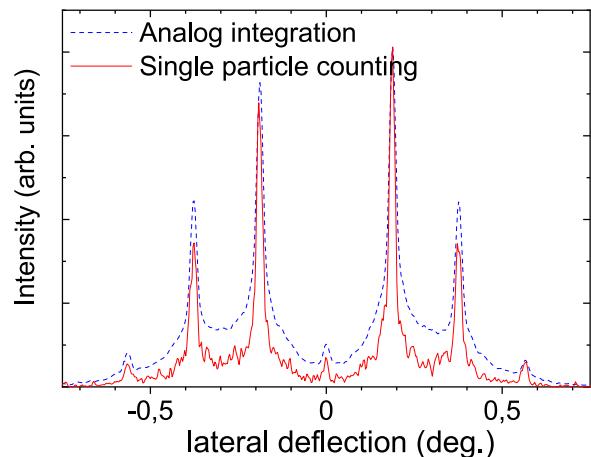


FIG. 6. Two diffraction profiles recorded on the Laue circle in similar conditions. The blue dashed curve corresponds to the standard analog integration mode, while the better contrasted red one is recorded in the single particle mode performed directly inside the camera.

10 frames/s without significant frame loss. The limitation is that the probability to have an overlapping spot should be negligible. This implies that the count rate within the surface of a spot is less than 5%–10% per frame.

The improvement in contrast and resolution is illustrated in Fig. 6, which compares the images recorded in the analog and single-particle counting mode. When analyzing the tiny spot of a low-intensity primary beam, the standard deviation $\sigma = 2.15$ pixel measured in the analog integration mode was reduced to $\sigma = 1.65$ in the photon counting mode, suggesting a contribution of around 1.5 pixel due to the size of a single-particle. This is fully consistent with the value of $\sigma = 1.6$ measured from a statistical analysis of a scattering image exposed to 100 ms where a few hundred individual impacts were visible.³⁵

VI. LARGE ANGLE SCATTERING

For beam and detector ports perfectly aligned, our detector³⁷ with 60 mm visible diameter and a distance to the target center corresponds to a maximum scattering angle of 1.5° . This latter could be doubled by giving an angle either of the beam or to the detector, but we have chosen to offer the possibility to explore larger scattering angles simply by putting commercial DN100CF hydro-formed bellows between the UHV chamber and the tube holding the detector (a DN63CF or DN50CF would have been large enough). The rotation takes place in the horizontal plane with an axis lying in the middle of the bellow, while the detector side sits on a 200 mm long linear rail with motor control and digital caliper. The camera is attached to the detector flange, and the maximum scattering angle is now close to 4° – 5° . Taking the maximum beam energy of 10 keV, this corresponds to an energy E_\perp of the motion perpendicular to the surface close to 100 eV, while the lowest energy E_\perp demonstrated was around 3 meV,²⁹ more than four orders of magnitude lower. With a helium projectile impinging LiF surface, elastic diffraction was observed until $E_\perp \sim 0.5$ eV, inelastic diffraction with well-resolved peaks was observed until $E_\perp \sim 1.5$ eV, and supernumerary rainbows corresponding to unresolved but coherent diffraction peaks was observed until $E_\perp \sim 4.5$ eV.⁴ It is likely that quantum effects such as the principal rainbow profile³⁸ persist above 5–10 eV, but its contribution may be hidden below instrumental resolution. These comparatively large values bridge the gap between GIFAD and all surface investigations performed with ions.³⁹ The minimum value of E_\perp was limited above 1 eV due to the image charge attraction before neutralization.⁴⁰ The relation between impact position (y, z) referred to the beam position and scattering angles (θ_y, θ_z) is given in Eq. (4) and depicted in Fig. 7; it is hardly more complex than with a straight tube but requires specific calibrations, as described in Fig. 13(c),

$$\begin{aligned} \theta_z &= a \tan \frac{z \cos^2 \theta_2 + a}{(z/2) \sin 2\theta_2 + a}, \\ a &= (L_2/2) \sin 2\theta_2, \\ b &= L_1 \cos \theta_2 + L_2 \sin^2 \theta_2 + L_2, \\ L_{\text{eff}} &= (L_1 \cos \theta_2 + L_2) / \cos(\theta_z - \theta_2), \\ \theta_y &= a \tan(y/L_{\text{eff}}). \end{aligned} \quad (4)$$

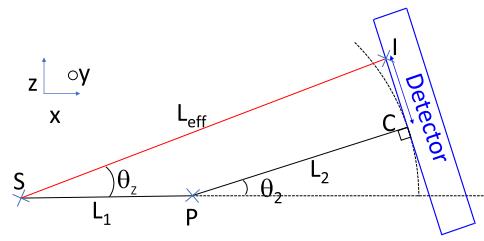


FIG. 7. The path to the detector is made of two sections (Fig. 2). A fixed tube of length L_1 and a tube of length L_2 connected by a bellow, allowing for a relative angle θ_2 . For each impact coordinate $z = IC$, the emission angles θ_y, θ_z from the surface are given by Eq. (4).

VII. EXPERIMENTAL METHOD

For complete data analysis, it is mandatory to know exactly where the direct beam is located and its exact shape. This usually requires two separate images: one where the direct beam is reasonably well centered so that the exact shape can be analyzed without distortion. One with the beam located close to an edge so that most of the detector is left free to analyze scattered particles. To avoid damaging the detector with a too intense beam, we have installed 1% and 10% transmission grids mounted on a translation stage before the detector. It is not mandatory when all the diaphragms reducing the beam divergence and intensity are set to a low value, but otherwise, the beam intensity can be significant. Even with this attenuated beam, the two images of the beam are recorded with a few ms exposure time and are typically recorded only once a day since most of the measurements are associated with variations of the target surface parameters, incidence angle, azimuth angle, or temperature. As usual, for data acquisition with cameras, the signal-to-noise ratio is improved by recording a reference file without the atomic beam by closing the valve just before the UHV chamber. We use the native software to drive the camera, and the background file is automatically subtracted from the one recorded with the atomic beam. This is particularly important if the atom count rate is low so that long exposures are needed. In this case, the nonuniform camera sensitivity and noise would slowly hide the signal. These last steps are not needed when the camera is operated in the single-particle detection mode, as described in Sec. V B. The data analysis is performed by a homemade software written in Borland C++, allowing for customized polar transform, as described in Ref. 9, and simple database access to the exact beam position and line shape associated with each image. When the interest is on the topology of the potential energy surface, strongly connected to the surface electronic density, only the elastic diffraction is of interest in each image. This corresponds to the intensity on the Laue circle of energy conservation clearly visible in Fig. 1. It is defined by $k_{in}^2 = k_{out}^2 = k_y^2 + k_z^2$. For convenience, this intensity is transferred onto a straight line by a polar-like transform,⁹ allowing for a direct comparison with the model surface topology using simplified^{8,13} or exact models^{1,41} to describe the quantum dynamics of the projectile. This representation on a straight line is also used to plot diffracted intensities as a function of time or angle of incidence or primary energy, as illustrated in Sec. VIII.

VIII. θ , ϕ , E , T , SCANS AND GROWTH MONITORING

The manipulator is motorized, and the control unit driving the motors is able to emit a synchronization signal to the camera so that automatic ϕ -scan and θ -scans can be programmed with small steps without close surveillance. For instance, a z -scan can be programmed to search the surface region that produces the best diffraction image. Figure 8 displays the evolution with the angle of incidence of the intensity recorded on a narrow slice around the Laue circle, showing a regular evolution of maxima and minima every second diffraction $\delta k_y = 2G_y$. This indicates a simple unit-cell structure with only one maximum and minimum per lattice unit. In addition, along z with a pseudoperiod δk_z indicating a corrugation amplitude $z_c \approx 2\pi/\delta k_z$, which hardly changes with the incident value of the angle of incidence $\propto k_z$. This almost constant corrugation amplitude was understood long ago with thermal atoms.^{42,43} The attractive van der Waals forces create a weak potential energy well with a depth D , increasing the incoming energy E_{\perp} to $E_{\perp} + D$. Of course, this is not an exact rule, and strong evolution of the corrugation was observed on simple systems due to the evolution from a single to a double maximum situation.^{44,45} For more complex unit-cells, the intensity modulation is also more complex, but simple optical models managed to link the observed features to the cell topology with surprising accuracy.^{6,9} During an energy scan, the angle of incidence is constant, and so is the radius of the Laue circle, while the Bragg angle or peak separation G_y along y is no longer constant. Figure 9 shows a typical increase of the mean scattering width at low energy, which was explained as a refraction effect due to the well-depth D .⁴¹ Figures 8 and 9 appear different, but the diffracted intensities recorded during an E -scan and θ -scan coincide when plotted as a function of the perpendicular energy $E_{\perp} = E \sin^2 \theta$,⁴¹ as predicted by the Axial Surface Channeling Approximation (ASCA) well established theoretically.^{46–50} Along each direction, the 3D potential energy landscape is replaced by its 2D average along this direction, and all methods developed to interpret TEAS elastic diffraction can be applied to GIFAD. Any given potential energy surface, either predicted by theory or simply guessed based on chemical considerations, can be compared with an

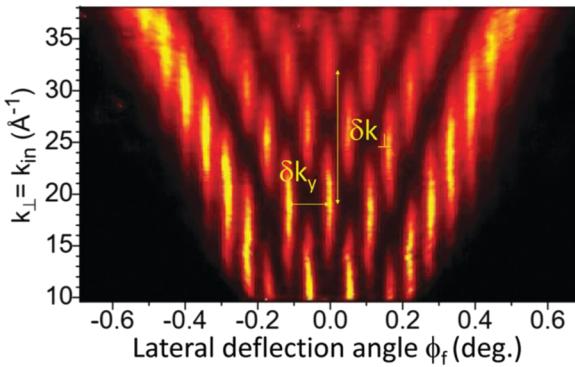


FIG. 8. During a variation of the incidence angle θ , the diameter of the Laue circle (Fig. 1) increases, but the spacing between diffraction orders stays fixed. Here, 500 eV Ne projectiles along with LiF[110]. Adapted with permission from Debiossac et al., J. Chem. Phys. Lett. 11, 4564 (2020). Copyright 2020 American Chemical Society.

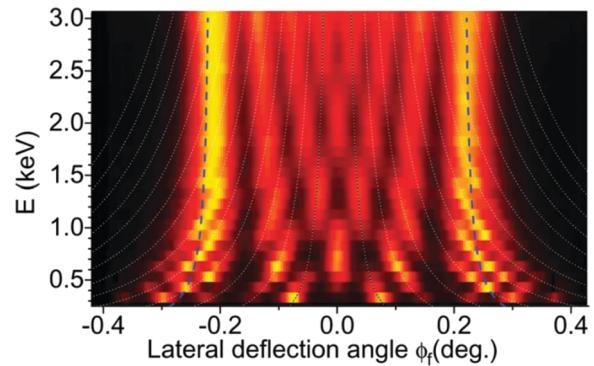


FIG. 9. During a variation of the primary beam energy E at $\theta = 0.57^\circ$, the diameter of the Laue circle is fixed, while the spacing between diffraction orders varies. The increase of the lateral scattering angle at low primary energy is due to attractive forces. Adapted with permission from Debiossac et al., J. Chem. Phys. Lett. 11, 4564 (2020). Copyright 2020 American Chemical Society.

experimental one using a quantum scattering calculation.^{46,47} Alternately, the potential energy surface fitting best to the data can be extracted from a close loop optimization with a fast algorithm for quantum dynamics.⁴¹ Returning to the experiment, one advantage of the E -scan compared with the θ -scan is that the surface illuminated by the beam does not change, and fine-tuning a 200 eV beam energy by only ~ 1 eV gives a sub-meV accuracy on E_{\perp} , as needed to explore bound state resonances.²⁹

Figure 10 displays a ϕ -scan, also called triangulation curve,⁵¹ where the width σ_{φ} of the lateral scattering profile is recorded during an in-plane rotation of the surface. Each peak, here repeated every 60° , corresponds to a channeling along a $\text{Al}_2\text{O}_3/\text{Ni}_3\text{Al}(111)$ surface low index direction.⁵² The peak amplitude corresponds to the illuminated section of the Laue circle in Fig. 1 or to the opening angle of

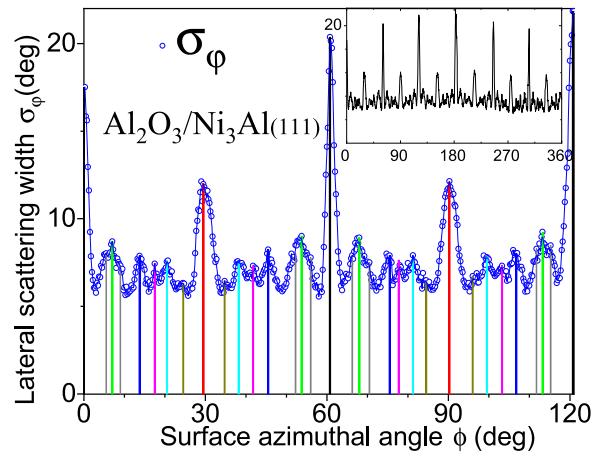


FIG. 10. Triangulation curve reporting the evolution of the mean width σ_{φ} of the scattering profile during a ϕ -scan or in-plane rotation with $\varphi = \arctan \frac{k_{f_y}}{k_{f_x}}$. The inset displays the evolution over a full turn. Several surface channeling directions are readily identified (in collaboration with A. Ouvrard⁵²).

the V-structure in Fig. 8 or to the distance between the vertical structures in Fig. 9. Theoretically, this width $\sigma_\phi = (\Sigma(\phi - \bar{\phi})^2)^{1/2}$ can be calculated from quantum mechanics, but, in practice, since all quantum contributions are averaged,^{53–55} the comparison with classical trajectory calculations¹⁶ readily provides a quantitative estimate of the surface structure. In practice, such a ϕ -scan is the first measurement after introducing a new sample. This can be achieved even with a beam resolution or with a surface coherence that does not allow for the observation of diffraction. After this step, diffraction can be investigated with high resolution along all directions where a peak is observed.

Finally, the simplest possible scan is a time evolution of the diffracted intensities at fixed positions of the target surface. This can be used to identify the surface reconstructions taking place at different temperatures⁵⁶ or to track the growth parameters under exposure to molecular beams from evaporation cells. Figure 11(a) shows pronounced oscillations of all measurable parameters of the diffraction images during a layer by layer, homo-epitaxial growth of GaAs/GaAs at 600 °C inside a MBE vessel at INSP.^{9,10} the mean scattering angle (θ_{out}), the width σ_θ , or the intensity on the Laue circle. These oscillations are directly related to the surface reflectivity, which decreases rapidly when ad-atoms are present on top of a new layer under grazing incidence. For the same system, Fig. 11(b) shows that at 480 °C, the growth oscillation is shifted by half a monolayer, revealing the well-documented phase transition from the more arsenic-rich $c(4 \times 4)$ reconstruction, changing to the $(2 \times 4)\gamma$ at the onset of growth.¹⁰ Note that similar results were obtained with RHEED or with keV ions at grazing incidence,^{57,58} outlining the dominant role played by ad-atoms on the surface specular reflectivity. Similarly to TEAS,^{42,43} the interest of GIFAD lies in the non-destructive behavior of atoms at low effective normal energy E_\perp and the absence of charging effect. In addition, there are not many techniques able to help online monitoring of the thin-film growth of fragile molecular layers.^{18,19,51} Note that the

contrast of the oscillations can be further improved by reporting only the elastic intensity,⁹ but this requires a more elaborate image processing.

IX. PROBING THE SURFACE QUALITY

Under grazing incidence, any protruding defect can severely perturb the projectile trajectory. This sensitivity appears at two different levels: the trajectory level discussed here and the macroscopic level as discussed at the end of this section. The elastic diffraction intensity approximately corresponds to the intensity sitting on the Laue circle visible in Fig. 1. It is very sensitive to surface defects at the trajectory level in the sense that probably a single protruding ad-atom along the trajectory is enough to push the atom away from perfect specular reflection. The length L of the trajectory can be evaluated from classical trajectories, but since diffraction is involved, the transverse (L_y) and longitudinal ($L_x = L_y/\theta$) coherence lengths are more relevant to defining the surface $S = L_x \cdot L_y$ needed for coherent reflection. Neglecting the energy dispersion, these are determined by the momentum spread associated with the angular spread of the beam $\delta k_\perp = k\delta\theta$. Taking into account the angle of incidence θ , the surface coherently illuminated is $S = (k^2 \delta\theta^2)^{-1}$. For 500 eV He and with our resolution of 0.1 mrad, this gives a surface $S \sim 5000 \text{ \AA}^2$ at 1° incidence. In principle, a lack of periodicity in this surface could ruin elastic diffraction. Since the longitudinal coherence L_x is $1/\theta$ longer than the transverse one L_y , it probably means that the mean distance between defects called surface coherence should be larger than $L_x \sim 200 \text{ \AA}$; otherwise, the elastic intensity will be reduced. On the macroscopic level, the surface illuminated by the beam is ϕ^2/θ , which is closer to a mm² for $\theta = 1^\circ$. It means that the diffraction signal can be used for online monitoring of the surface coherence length of up to several hundred Å and that the diagnostic applies over a significant surface around 1 mm². Macroscopic defects such as tilt and twist surface mosaics⁵⁹ and wafer curvature induced by surface tension can be tracked *in situ* and online at the mrad level. More work is needed to identify specific signatures associated with other types of defects such as specific terraces.

X. COMPLEMENTARY EQUIPMENT

A. The ion beam

If no gas is fed in the charge exchange cell, the intense ion beam can be used for surface analysis or surface preparation by Grazing sputtering.^{39,60} For the LiF surface, the best surface quality was obtained by cleaving the surface (see, e.g., Ref. 4). The situation is opposite for metals, where the diffraction on the Ag(110) surface could be observed only after repeated cycles of sputtering and annealing.^{11,61}

The presence of a keV ion or atom beam is enough to count the number of deposited layers during growth by tracking the intensity oscillation of the quasi-specular beam.^{10,57,62} This analysis in terms of reflection coefficient or reflectometry is almost identical for atoms or ions. Both have comparable trajectories above the surface; however, due to image charge attraction, the minimum impact energy is above 1 eV,^{40,63,64} making elastic scattering very unlikely. When the energy perpendicular to the surface E_\perp is close to or larger than 10 eV, the projectile penetrates the electronic

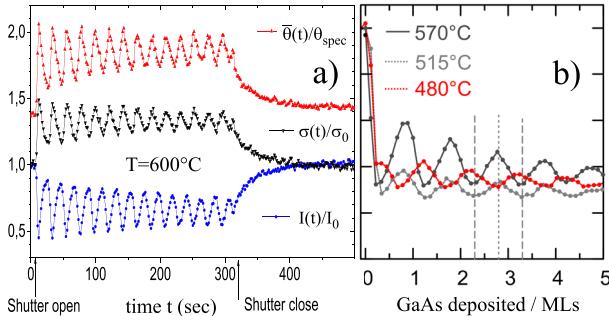


FIG. 11. The imaging detector allows for simple tracking of the number of deposited layers during a layer by layer growth of GaAs/GaAs. Pronounced oscillations are visible for the intensity $I(t)$ around the Laue circle, the mean polar scattering angle $\theta_{out}(t)$, and the polar scattering width $\sigma_\theta(t)$ (a). Adapted with permission from M. Debiossac and P. Roncin, *Appl. Surf. Sci.* **391**, 53–58 (2016). Copyright 2016 Elsevier B.V. The classical nature of these parameters ensures a robust behavior. (b) At $T = 480$ °C, the completion of the first layer is delayed by half a growth oscillation due to an initial surface reconstruction consuming Ga (b). Adapted with permission from Atkinson *et al.*, *Appl. Phys. Lett.* **105**, 021602 (2014). Copyright 2014 AIP Publishing LLC.

density of the target at distance close to 1 Å and secondary electrons are emitted. By tracking the secondary electron yield during an azimuthal scan, some surface structure parameters, such as the direction of the low index axis, can be determined by ion beam triangulation.⁶⁵ This is equivalent to the ϕ -scan or atom beam triangulation described above, where the increased lateral scattering width identified the channeling direction due to the repeated deflections on the walls of the valley. Here, the enhanced electron emission is partly due to the comparatively long zig-zag trajectories inside the valleys. This correlation between selected trajectories associated with electron emission can be identified by coincidence experiments using time-resolved detectors,^{25,33,37,66} allowing trajectory-dependent electron emission to be identified.^{67,68} By pulsing the ion-beam, the projectile energy loss can be measured, allowing for the identification of surface excitations, such as ballistic phonons,^{69,70} optical phonons^{40,71} or excitons and trions.^{72,73} Orienting the surface to large angle of incidence opens the field of ion beam analysis, such as time of flight analysis of the ejected particle⁷⁴.

B. The webcam and perpendicular laser

The UHV chamber offers a DN100CF flange directly facing the target surface. When unused, it is closed by a window flange where we have installed a webcam and a miniature red laser. The red laser is shifted by ≈ 2 cm from the center and directed to intercept the surface around its center. If the target surface is, indeed, parallel to the flange, the reflected beam also lies 2 cm from the center but opposite to the primary spot. During an azimuthal rotation, the reflected spot describes a circle with a center located at $2\theta_{laser}$ and a radius 2τ , where τ is the misalignment of the surface normal with respect to the rotation axis and θ_{laser} is the arbitrary angle of the laser with the mean surface normal. Due to the 30 cm long path between the surface and the window, the position of the spot reflected from the surface is easily tracked by using the camera. It can be used to follow the target azimuthal angle online and to correct for the possible tilt τ .⁷⁵

C. The co-axial laser

The alignment procedure of all the diaphragms and differential pumping holes is easier with the miniature X, Y, θ_X, θ_Y platform attached directly to the EX05 ion source (Sec. II). As detailed in Sec. V B, when a single MCP is used together with a phosphor screen without an aluminum over-layer, there is enough light passing through the (unbiased) MCP to be imaged by the camera, allowing for quantitative intensity optimization both in vacuum or at atmospheric pressure.

This allows for a very simple pre-positioning of the target surface into the beam and direct measurement of the angle of incidence, which can be different from a crystal plane in the case of miscut. In this case, the atoms would reveal the crystal plane, while the light would reveal the mean surface plane.⁷⁶ Since the laser impact on the surface is visible and can be captured by using a webcam, a mark can be “taped” on the picture of the target surface where a poor or good diffraction is observed (Fig. 12).

This is also convenient to calibrate the diaphragms and slits sizes and positions by analyzing the observed Airy pattern in

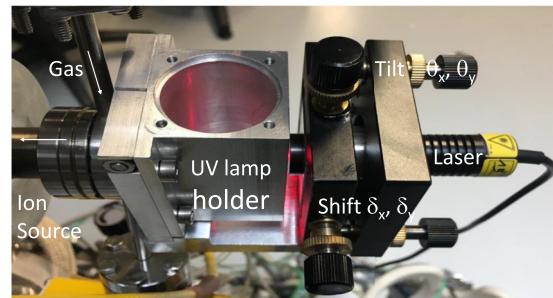


FIG. 12. The colinear laser is held on a miniature X, θ_X, Y, θ_Y platform hooked to a view-port at the gas injection side of the ion source.

Fig. 13(a). It also allows for an accurate angular calibration procedure. A shortcoming attached to our detector is that the calibration is very sensitive to the exact zoom value of the camera lens. This can later be optimized online, with the primary beam to produce a minimum spot size. It is sometimes useful to check the exact overall angular scale depending on actual distances and zoom values, which is not always easy to determine. We have used a commercial transmission electron microscope grid called mesh-300 with a $d = 84\mu\text{m}$ periodicity, point-welded on the backside of the manipulator's head so that it can be inserted into the laser beam to generate bright spots with $\theta_{Bragg} = \lambda/d$ in rad. The diffracted spots of Fig. 13(c) allow for the simple calibration of the detection system, whatever the optical elements and actual detector position or orientation.

It also has nice educational potential as many undergraduate students are surprised that visible light diffracts on the collimating diaphragms but, apparently, not on the surface, while keV atoms do exactly the opposite. Note that using a single free-standing graphene layer and a geometry demonstrated with highly charged ions,⁷⁷ diffraction of atoms through graphene was predicted.⁷⁸ It also offers an opportunity to define the Airy function ubiquitous in wave mechanics, from atmospheric rainbows to atomic collisions.^{79,80} Here, in atomic diffraction at surfaces, the Airy function and its companion Bessel function can be derived by the semi-classical intensity modulation observed in diffraction from the simple lattice unit and known as supernumerary rainbows^{3,4,38,62,81} visible in each line of Figs. 8 and 9.

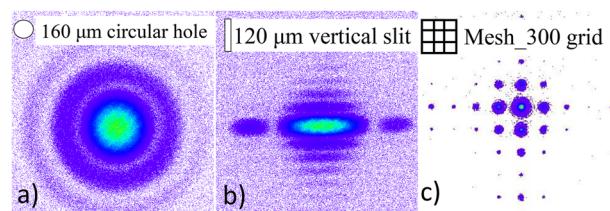


FIG. 13. (a) Laser diffraction through $\varnothing_2 = 160\mu\text{m}$ and (b) through a $120\mu\text{m}$ wide horizontal slit. (c) Diffraction through the $84\mu\text{m}$ pitch of a TEM grid at target position. The Bragg angles are used for *in situ* angular calibration.

XI. CONCLUSIONS

The setup presented here is a versatile research setup. A more compact design of the source section integrating the Wien filter and neutralization cell is possible but has not been attempted. The energy E_{\perp} associated with the motion perpendicular to the crystal axis is a key parameter that governs the minimum distance of approach to the surface. The setup allows for the operation with atoms from $E_{\perp} \approx$ a few meV up to a few 100 eV, opening a direct connection with the keV ion-surface community⁸² and all its surface sensitive techniques in continuity with the more penetrating, higher energy projectiles of the ion-beam-analysis community.⁸³ We have focused on systematic errors associated with small angles. These are related to the control of the primary beam, with the target positioning, and to the mechanical, optical, and electronic aberrations in the measurement method. The combination presented here of a single MCP with a phosphor screen imaged at medium distance by a high aperture lens corrected by software and using a high efficiency CMOS camera operated in the single impact counting mode provide accurate positions and intensities. It is very close to an ideal detector with high resolution and, most important, an excellent integral and differential linearity. *A priori*, the conditions where GIFAD could be helpful are the growth of ultra-thin or fragile layers and severe electromagnetic environment. It could also help production of high-quality surfaces, i.e., with very large coherence length. In both cases, the ability of GIFAD to provide an online handle to optimize the growth parameters, such as surface temperature and evaporation rate, could be decisive. In many situations, a very simple numerical treatment of the diffraction image can provide “on the fly” quantitative parameters, such as the reflectivity, the scattering width, and the elastic scattering ratio.

So far, for molecular layers, no detailed topology could be extracted directly from the diffraction pattern; however, the directions where the molecules tend to align are immediately revealed by triangulation curves,⁵¹ such as the one displayed in Fig. 10, and these can be directly compared with classical calculations to select the most appropriate structural model.⁸⁴ The evolution of the reflectivity with time also allows for the identification of the structural change interpreted as a liquid to solid phase transition occurring when the density of highly mobile molecules at the surface is high enough¹⁹ as could be investigated by STM.^{85,86}

So far, fast ion diffraction has never been observed probably because ions interact too strongly with the surface electrons or with the ions via excitation of optical phonons in the case of insulators.^{71,87} Diffraction patterns were observed with primary ions but associated with scattered atoms that have neutralized far from the surface so that they impact the surface as neutral atoms.⁸⁸

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Peng Pan: Investigation (supporting). **Jaafar Najafi Rad:** Investigation (equal). **Philippe Roncin:** Formal analysis (lead); Investigation (equal); Project administration (lead); Writing – original draft (lead); Writing – review & editing (lead).

DATA AVAILABILITY

Data available on request from the authors.

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